

PII S0160-4120(98)00020-8

## ASSESSMENT OF AIR QUALITY IN PARIS BY PERSONAL MONITORING OF NONSMOKERS FOR RESPIRABLE SUSPENDED PARTICLES AND ENVIRONMENTAL TOBACCO SMOKE

K. Phillips, M.C. Bentley, and D.A. Howard

Department of Air Quality Monitoring, Covance Laboratories Ltd. (formerly Corning Hazleton Europe), Harrogate, England

G. Alván

Karolinska Institute, Department of Medical Laboratory Sciences and Technology, Division of Clinical Pharmacology, Huddinge University Hospital, Huddinge, Sweden

*E1 9709-182 M (Received 30 August 1997; accepted 25 December 1997)*

Exposure to respirable suspended particles (RSP), environmental tobacco smoke (ETS) particles, nicotine, and 3-ethenylpyridine (3-EP) was assessed in Paris for 222 subjects during March and April 1995. Personal monitors were worn over a 24-h period, each subject providing a saliva sample for cotinine analysis both prior to and following the monitoring period. Comprehensive lifestyle questionnaires were also completed before and after the monitoring period. The study comprised housewives in one group, primarily for assessing exposures in the home, and office workers in a second group to assess exposures in the workplace. A single personal monitor was worn by each housewife, while employed subjects wore one monitor at work and a separate monitor at home and elsewhere. Based on median 24-h time-weighted average exposures, the most highly exposed subjects to RSP, ETS particles, nicotine, and 3-EP were office workers living with smokers. Additionally, based upon nicotine exposures, subjects who were also employed in locations where smoking was allowed were the most exposed. Based upon median levels, housewives living in nonsmoking households would be exposed to 1 cigarette equivalent per y or less, compared with between 1.2 and 3 cigarette equivalents per y for housewives living in smoking households. Subjects living and working with smokers had the highest median saliva cotinine level of  $1.6 \text{ ng mL}^{-1}$ . Using a cut off level of  $25 \text{ ng mL}^{-1}$  to indicate active smoking, up to 4.7% of the subjects were found to have misreported themselves as nonsmokers. ©1998 Elsevier Science Ltd

### INTRODUCTION

Proposals for setting ambient air quality standards for specific pollutants throughout the European Community are due for publication in 1997. For particles, the 24-h mean will be  $50 \mu\text{g m}^{-3}$  (to be achieved 98% of the time). Threshold values will also be set for major industrial and vehicular pollutants such as  $\text{SO}_2$  and  $\text{NO}_2$ . Studies investigating the health impacts of airborne

particles are currently focusing on the smaller particle sizes rather than grouping all particles smaller than 10 microns ( $\text{PM}_{10}$ ). The United Kingdom's Expert Panel on Air Quality Standards (EPAQS) recommended in 1995 that a  $\text{PM}_{2.5}$  standard be considered by the year 2000. When lung damage and other health problems associated with fine particles are more fully

understood, the measurement of fractions smaller than  $PM_{10}$  may have more significance. Airborne particles emitted from natural sources vary in size and are not controllable whilst man-made particle emissions are often smaller in diameter and can normally be controlled with additional cost implications. Reducing the overall standard to  $PM_{2.5}$  will inevitably result in legislative problems.

Paris was the fourth successive major European city studied by these authors on air quality following investigations in Stockholm (Phillips et al. 1996), Barcelona (Phillips et al. 1997a), and Turin (Phillips et al. 1997b). The study was carried out during March and April 1995 with personal monitoring taking place over a 24-h period. In addition, volunteers self-reported activities using diaries and questionnaires. Saliva samples were taken for cotinine analysis before and after the 24-h period. The particle fraction collected was  $PM_{3.3}$ , suitable for capturing particles produced during smoking. Exposures to respirable suspended particles (RSP), environmental tobacco smoke (ETS) particles, nicotine, and 3-ethenylpyridine (3-EP) were evaluated in homes and workplaces. ETS particles were measured using ultraviolet absorbing particulate matter (UVP), fluorescing particulate matter (FPM), and solanesol related particulate matter (SolPM). Other recent studies used similar methodologies (Sterling et al. 1996; Heavner et al. 1996; Jenkins et al. 1996; Baek et al. 1997).

In the context of this publication, the term "exposure" can be taken to mean the "potential inhaled quantity", calculated as the product of the encountered concentration, the length of time subjected to such concentration, and the breathing rate maintained throughout the defined period. Exposures are also quoted in terms of cigarette equivalents (CE), these having been calculated in relation to the mainstream particle and nicotine yields of typical French cigarettes. The values, 13 mg particles and 1.0 mg nicotine, were calculated from the mean yields of the top six selling cigarette brand-types in France.

Data generated from personal monitoring studies are not normally distributed and, for this reason, median values for RSP and ETS exposures are reported in preference by many investigators. Some authors have reported arithmetic or geometric means and, to enable comparison with these findings, these parameters have been reported alongside median values for each data set. Also reported are 10th and 90th percentile values, also referred to as the lower and upper deciles, respectively, as a more appropriate indication of the

range of values than the minimum and maximum. ETS particle concentrations, corresponding exposure calculations, and comparisons between subject groups and Cells have been based primarily upon SolPM estimates. These are generally considered to give the closest representation of actual ETS particle concentrations, but, as indicated previously (Phillips et al. 1997a; 1997b), it was considered appropriate to report FPM estimates alongside those for SolPM.

The main objective of this study was to determine personal exposures of nonsmoking inhabitants in Paris over a 24-h period, and to assess and compare these exposures in the workplace and at home. Homes and workplaces were classified as either "smoking" or "nonsmoking". The following definitions were chosen for consistency across different countries investigated: homes were classified as "smoking" if a smoker of cigarettes, pipes, or cigars was resident and also normally smoked within communal areas of the household. The smoking "status" of a workplace was defined by the absence/presence of smoking co-workers within 30 m of the volunteer's workstation and no consideration was taken regarding the employer's rules or local authority regulations.

## METHODS

### *Recruitment of subjects*

The Mosaic geodemographic classification system, as utilised by marketing companies and used previously by these authors (Phillips et al. 1996), was not available at the time of recruitment in Paris. Instead, a local segmentation system, which classified the inhabitants of Paris into "clusters" based upon the same principle as Mosaic, was employed. The sample was chosen to be compliant with the following criteria:

- 1) All subjects were nonsmokers living within 15 km from the city centre of Paris.
- 2) A third in each of the three age groups 20-34, 35-49, and 50-64.
- 3) Equal percentage distribution in geodemographic "clusters" as for the population within 15 km of the city centre.
- 4) Subjects were distributed between six "Cells" as indicated in Table 1, Cells 3-6 being targeted at office workers.

The volunteers in this study were recruited using randomly selected telephone numbers from files created in accordance with the above criteria. Initial telephone contact was performed by Teleperformance, a large opinion research bureau resident in Paris, at which

Table 1. Cell categorisation by home and workplace status (Paris).

Cell	Study type	Smoking status		Planned number
		Home	Work	
1	Single monitor	Smoking	-	55
2	Single monitor	Nonsmoking	-	40
3	Dual monitor	Smoking	Smoking	45
4	Dual monitor	Smoking	Nonsmoking	30
5	Dual monitor	Nonsmoking	Smoking	40
6	Dual monitor	Nonsmoking	Nonsmoking	30

point prospective study participants were screened to confirm their eligibility to take part. This screening included questions concerning age, current and previous smoking status, other nicotine product use, and employment status. Suitable volunteers were then given an appointment to attend an information/training session organised at the Hotel Mercure Tour Eiffel in Paris.

Once Mosaic classification has been developed for France, a retrospective comparison of the present study sample with the population of Paris may be possible.

#### The monitoring session

Volunteers were required to wear a personal monitor, designed to collect particulate and vapour phase components present in the air close to the subject's breathing zone (Ogden et al. 1996), over a period of 24 h. RSP and ETS particles were collected onto a Fluoropore membrane filter and vapour phase components, nicotine and 3-EP, were adsorbed onto XAD-4 resin beads. The personal monitoring methodology was described previously by these authors (Phillips et al. 1996) and consisted briefly of the following:

*Initial visit to the study centre:* Subjects were issued personal monitors and diaries/questionnaires for recording exposures and observations over the 24-h collection period. Full instructions regarding the use of the monitoring equipment and how to complete the study diaries and questionnaires were provided by means of a video presentation and the assistance of locally recruited translators to provide additional clarification where necessary. Nonworking subjects recruited for participation in Cells 1 and 2 (housewives) were provided with a single personal monitor for use over the collection period (single monitor study). Working subjects recruited for participation in Cells 3 to 6 were provided with two personal monitors for use over the

same period (dual monitor study). Subjects were required to provide a saliva sample prior to the monitoring period (pre-sample).

*Final visit to the study centre:* Following completion of the 24-h monitoring period, subjects were required to return their personal monitors and associated documentation to the study centre. Subjects also provided a second saliva sample (post-sample) and completed a "last visit" questionnaire.

#### Analytical procedures

All analytical procedures were validated and were fully described previously by these authors (Phillips et al. 1996). In this study, the following analytes were determined:

*RSP*—using a gravimetric procedure (Ogden et al. 1990).

*Saliva cotinine*—using a radioimmunoassay procedure (Van Vunakis et al. 1987; Davis and Stiles 1993).

*Nicotine and 3-EP*—using a capillary gas chromatographic procedure (Ogden et al. 1989).

*Estimation of ETS particles (three methods)*—using high performance liquid chromatography (HPLC) procedures to determine the ultraviolet absorbance (UVP), FPM, or SolPM content of methanolic filter extracts (Ogden et al. 1990). The factors used in this study to convert instrument responses into an equivalent concentration of ETS particles were 36 (SolPM), 38 (FPM), and 6.9 (UVP) as determined by Nelson et al. 1997.

The limits of quantification (LOQ) for these analyses are presented in Table 2. Any data below the LOQ were assigned a value of  $\frac{1}{2}$  LOQ for statistical analyses.

Table 2. Limits of quantification for the analytical methods according to collection period (Paris).

Measurement	Collection period		
	24 h	15.04 h*	8.23 h**
Respirable suspended particles (RSP) <sup>†</sup>	9.9 $\mu\text{g m}^{-3}$	16 $\mu\text{g m}^{-3}$	29 $\mu\text{g m}^{-3}$
ETS particles measured by UV (UVPM) <sup>†</sup>	0.42 $\mu\text{g m}^{-3}$	0.67 $\mu\text{g m}^{-3}$	1.2 $\mu\text{g m}^{-3}$
ETS particles measured by fluorescence (FPM) <sup>†</sup>	0.10 $\mu\text{g m}^{-3}$	0.16 $\mu\text{g m}^{-3}$	0.29 $\mu\text{g m}^{-3}$
ETS particles measured by solanesol (SolPM) <sup>†</sup>	0.22 $\mu\text{g m}^{-3}$	0.35 $\mu\text{g m}^{-3}$	0.64 $\mu\text{g m}^{-3}$
Nicotine <sup>††</sup>	0.09 $\mu\text{g m}^{-3}$	0.14 $\mu\text{g m}^{-3}$	0.25 $\mu\text{g m}^{-3}$
3-Ethenylpyridine (3-EP) <sup>††</sup>	0.09 $\mu\text{g m}^{-3}$	0.14 $\mu\text{g m}^{-3}$	0.25 $\mu\text{g m}^{-3}$
Saliva cotinine	1.0 ng mL <sup>-1</sup>	-	-

\* Mean time spent outside the workplace for working subjects in Paris.

\*\* Mean time spent at work for working subjects in Paris.

<sup>†</sup> Calculated assuming a flow rate of 1.72 L min<sup>-1</sup> through the Fluoropore filter.<sup>††</sup> Calculated assuming a flow rate of 0.8 L min<sup>-1</sup> through the XAD-4 tube.

Table 3. Age and sex distribution for study subjects (Paris).

Cell	Sex			Age range				Overall total
	Males	Females	Unknown	20-34	35-49	50-64	>64	
1	1	48	2	24	15	12		51
2		44		13	16	14	1	44
3	12	28	5	24	13	8		45
4	4	9		6	5	2		13
5	27	27	5	30	16	13		59
6	2	7	1	6	1	3		10
Single monitor total	1	92	2	37	31	26	1	95
Dual monitor total	45	71	11	66	35	26		127
Overall total	46	163	13	103	66	52	1	222

Cell 1: smoking household; Cell 2: nonsmoking household; Cell 3: smoking household/smoking workplace; Cell 4: smoking household/nonsmoking workplace; Cell 5: nonsmoking household/smoking workplace; Cell 6: nonsmoking household/nonsmoking workplace.

## RESULTS AND DISCUSSION

### Subject selection

Of the 240 subjects initially recruited for the study, 5 were excluded because they failed to collect their samples, 7 subjects were excluded after they admitted to being smokers during their initial visit to the study centre, and 4 subjects were excluded because their saliva cotinine levels were above the selected threshold (25 ng mL<sup>-1</sup>) for nonsmokers. Two subjects were excluded due to the absence of saliva cotinine data required to confirm their nonsmoking status.

The age and sex distributions of the remaining 222 subjects who successfully completed the study are presented in Table 3. Figure 1 shows that the sex dis-

tribution for employed subjects was slightly biased towards female subjects, although it should be noted that gender information for approximately 9% of the subjects was not provided by Teleperformance. Figure 2 shows the spread of subjects by age group in both single and dual monitor studies, together with a combined distribution for all recruited subjects. There was a bias towards the younger age groups, particularly for the dual monitor study, from the targeted 33% in each group. This may be attributable to difficulties in recruiting workers in the 50-64 y age group. One subject whose age fell outside the specified age range (> 64) was included in the study so that much needed data on ETS exposure in the home could be obtained.

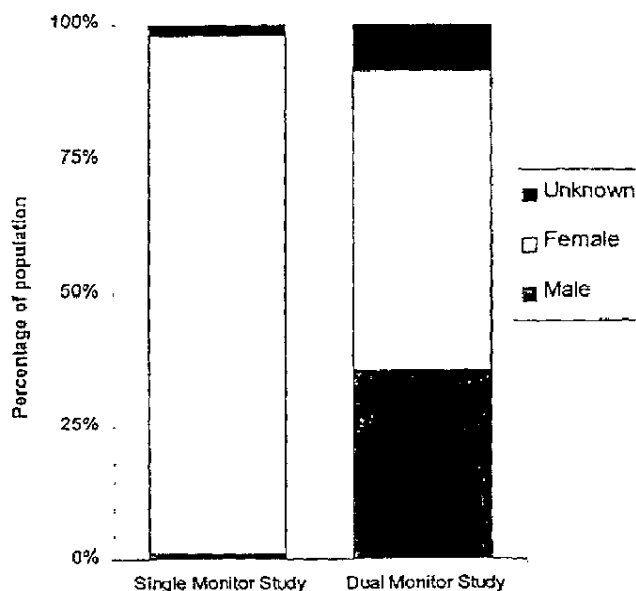


Fig. 1. Sex distributions for volunteers on the single and dual monitor studies within the Paris recruitment area.

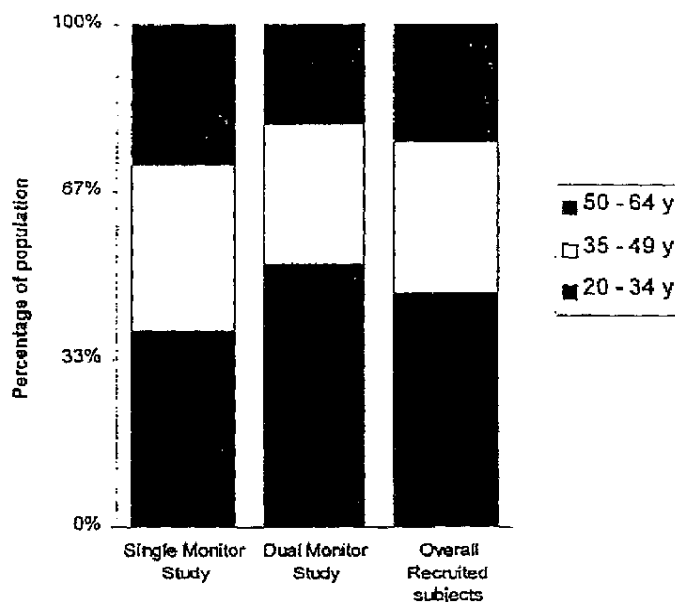


Fig. 2. Age range distributions for volunteers overall and for each study (Paris).

The participants were restricted to a choice of 12 occupations from which to select and provide their answers on the last visit questionnaire. Table 4 lists these occupations and the answers that were provided by 126 of the 127 subjects wearing the workplace monitor in this study.

#### *Weather and pollutant information*

Detailed information about the weather conditions during the course of the study were obtained from Météo France, the local meteorological office in Paris, and the levels of certain airborne pollutants were

Table 4. Occupations of recruited working subjects (Paris).

Occupation	Number of responses
Administrative/secretarial	33
Building/construction	3
Education	12
Engineering	9
Government agency (civil service)	16
Legal/financial	10
Hotel/restaurant/leisure industry	4
Medical	4
Other	20
Wholesale/retail	2
Science/computing	10
Supply industry	1
Transportation/haulage	2
Total	126

obtained from AirParif, an organisation responsible for air pollution monitoring in the Paris area. The study was carried out during March/April 1995 with daily mean temperatures over this period varying from a minimum of 5.6°C to a maximum of 15.3°C. A maximum daily rainfall of 5.2 mm was recorded with rain falling on 4 d of the study period. Mean daily wind speeds varied between 0.8 and 2.8 m s<sup>-1</sup> and maximum and minimum relative humidities of 94% and 31%, respectively, were recorded. Concentrations of particles, NO, NO<sub>2</sub>, SO<sub>2</sub>, and O<sub>3</sub> were also obtained from up to 10 monitoring stations situated around the Paris area. Daily mean NO<sub>2</sub> concentrations varied from 30 to 140 µg m<sup>-3</sup> and were in excess of 100 µg m<sup>-3</sup> on at least 5 d of the study period, when the air quality based upon UK bandings could be described as poor. Particle concentrations, expressed as daily means, varied from 90 to 1171 µg m<sup>-3</sup> with an overall average of 446 µg m<sup>-3</sup> during the study period.

The information regarding weather and pollutants is provided purely as a point of reference. No attempt was made to correlate this information with the personal monitoring data or with similar information from other cities.

#### *Smoking status and misclassification*

Saliva cotinine levels were determined in order to verify that recruited subjects had correctly reported themselves as nonsmokers. Various threshold levels above which subjects would be classified as smokers

have been suggested and include 10 ng mL<sup>-1</sup> (Etzel 1990), 15 ng mL<sup>-1</sup> (McNeill et al. 1987), 30 ng mL<sup>-1</sup> (Lee 1987), and more recently 100 ng mL<sup>-1</sup> (Sterling et al. 1996). In this study, 25 ng mL<sup>-1</sup> (maximum of pre- and post-levels) was used, as chosen and described previously by these authors (Phillips et al. 1994), as a suitable cut-off level. Using this threshold, one subject with a level of 27 ng mL<sup>-1</sup> was assumed to be an occasional smoker and a further three subjects, with levels between 122 and 213 ng mL<sup>-1</sup>, were assumed to be regular smokers and were excluded from the study.

Seven subjects were excluded from the study after admitting to being smokers on the first visit questionnaire, including users of pipes and/or cigars. One of these subjects had saliva cotinine levels in excess of 100 ng mL<sup>-1</sup>, indicative of a regular smoker. The remaining six subjects, whose levels varied between 0.5 and 3.9 ng mL<sup>-1</sup>, may have abstained from smoking for several days before the study started and/or were occasional smokers. Alternatively, they may have misreported their smoking status or were active smokers who poorly metabolise nicotine (Cholerton et al. 1994; Benowitz et al. 1995).

In this study, the subjects were required to confirm they had been nonsmokers for more than six months and no attempt was made to differentiate between "non" and "never" smokers. Various criteria can be used to assess the rate at which recruited subjects misreport their smoking status. A detailed discussion of smoking status misclassification was published recently by Ogden et al. (1997) and provides data from a large study in the U.S. Depending upon the criteria used, the rate at which the subjects misclassified their smoking status in this study ranged between 1.8% (4 from 226) and 4.7% (11 from 233). This compares with between 2.7% and 5.3% for Stockholm, between 10.5% and 17.8% for Barcelona, and between 1.6% and 6.5% for Turin, the values having been estimated in an identical way.

Etzel's review (1990) of the use of saliva cotinine for this purpose suggests that subjects with cotinine levels between 10 and 100 ng mL<sup>-1</sup> may be classified as infrequent smokers, and, had we selected 10 ng mL<sup>-1</sup> as the cut-off level, 4 more subjects would have been rejected. Delfino et al. (1993) rejected only 3 out of 251 subjects using a cut-off of 20 ng mL<sup>-1</sup>, whereas Sterling et al. (1996) could have rejected 2 out of 25 had they used a similar threshold level instead of 100 ng mL<sup>-1</sup>. In a study of cardiovascular risk factors in 5115 young adults (CARDIA), Wagenknecht et al.

Table 5. Correlation coefficients for ETS 'markers' using all data (Paris).

"Y" data	vs	"X" data	Data pairs	R-squared	Gradient	Intercept
FPM		UVP	342	0.833	1.040	-0.31
SolPM		UVP	340	0.826	1.208	-4.12
SolPM		FPM	340	0.651	0.939	-1.23
3-EP		Nicotine	333	0.802	0.379	0.11
FPM		Nicotine	329	0.451	7.216	4.12
SolPM		Nicotine	327	0.541	9.062	0.20
UVP		Nicotine	330	0.589	7.188	4.07
SolPM		3-EP	327	0.612	22.627	-1.75
FPM		3-EP	329	0.519	18.359	2.47
Post-cotinine		Nicotine*	202	0.196	0.539	0.90
Post-cotinine		SolPM*	209	0.103	0.025	1.11
Post-cotinine		FPM*	210	0.073	0.021	1.11

\* Time-weighted average concentrations correlated for working subjects (Cells 3 to 6).

Table 6. Correlation coefficients for ETS 'markers' using only data greater than the LOQ (Paris).

"Y" data	vs	"X" data	Data pairs	R-squared	Gradient	Intercept
FPM		UVP	340	0.832	1.040	-0.32
SolPM		UVP	246	0.817	1.233	-4.87
SolPM		FPM	246	0.624	0.932	-0.79
3-EP		Nicotine	204	0.767	0.355	0.21
FPM		Nicotine	314	0.444	7.172	4.29
SolPM		Nicotine	235	0.512	8.858	1.37
UVP		Nicotine	314	0.583	7.143	4.24
SolPM		3-EP	183	0.575	23.432	-2.71
FPM		3-EP	202	0.447	17.696	3.51

(1992) found a misclassification rate of 4.2% based on a serum cotinine cut-off of 14 ng mL<sup>-1</sup>. They classified nonsmokers as subjects having reported smoking less than 5 cigarettes per week for the previous 3 months. An exclusion rate of 4.2%, using a 15 ng mL<sup>-1</sup> saliva cotinine discrimination level, was also found on a recent personal air monitoring study (Jenkins et al. 1996) of 1564 subjects in the United States and misclassification rates of between 2.81% and 4.01% were estimated in a study of 900 married females in the U.S. (Ogden et al. 1997). In this study, saliva cotinine threshold levels of 35 and 106 ng mL<sup>-1</sup>, calculated according to the U.S. Environmental Protection Agency (USEPA) definitions, were used to differentiate between occasional and regular smokers, respectively.

#### Comparison of "markers" for estimating ETS concentrations

The correlation and best fit line coefficients between various analytes used to assess ETS levels in this study are listed in Table 5. Table 6 depicts the same information following the removal of data pairs where either analyte was below the LOQ. The cumulative frequency distributions for all analytes are presented in Figs. 3 and 4.

A good correlation ( $R^2=0.833$ ) between ETS particle measurements made using UVP and FPM methods was evident (Table 5), with a gradient and intercept indicating close similarity between the results. This was also reflected in Fig. 4, where near identical ETS particle distributions using both UVP and FPM

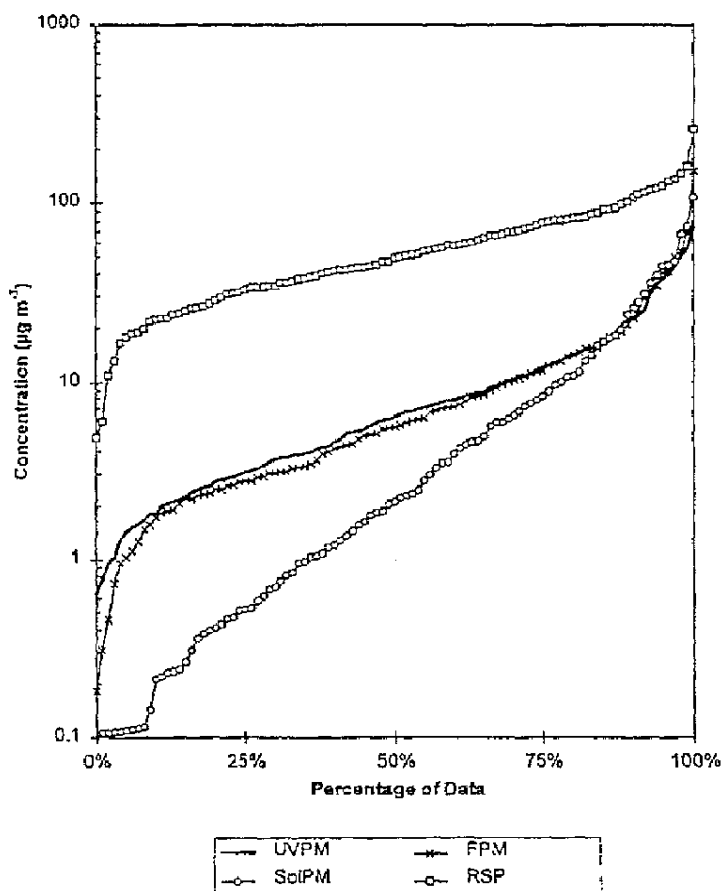


Fig. 3. Cumulative frequency distributions of average 24-h particulate matter concentrations (Paris).

estimates were observed. The correlations of SolPM measurements compared with either UVPM ( $R^2=0.826$ ) or FPM ( $R^2=0.651$ ) estimates were still good, although gradient and intercept values indicated that ETS particle concentrations determined using SolPM measurements gave higher values than for UVPM or FPM estimates above a certain concentration. This crossover point can be seen in Fig. 3 and is equivalent to an ETS particle concentration of approximately  $25 \mu\text{g m}^{-3}$  and may indicate the possibility of SolPM overestimating ETS particle concentrations at high levels. This phenomenon was observed and discussed previously by these authors (Phillips et al. 1997a) and was tentatively attributed to the factors used for calculating particulate concentrations.

It was also noted that 27% of the SolPM results were below the LOQ compared with 0.3% and 0.9% for FPM and UVPM, respectively. Baek et al. (1997), studying air quality in Korean homes, offices, and restaurants, similarly reported a number of SolPM esti-

mates higher than the corresponding FPM/UVPM results together with a high percentage of SolPM estimates below the LOQ. In this study in Paris, of the samples where the SolPM estimates were below the LOQ, 81% had measurable nicotine concentrations in the range  $0.07\text{--}2.0 \mu\text{g m}^{-3}$  (median  $0.20 \mu\text{g m}^{-3}$ ). It is possible that this apparent lack of sensitivity for SolPM determinations is due to the specificity of solanesol as a marker for ETS particles since the presence of nicotine, particularly at low levels, may be attributable to desorption from various surfaces in the absence of ETS particles (Nelson et al. 1990). However, in environments subjected to recent or active smoking, Eatough (1993) concluded that "the concentration of gas phase nicotine underestimates exposure to the particulate phase of environmental tobacco smoke constituents" due to the faster decay characteristics of nicotine. It is possible, therefore, that nicotine determinations can both overestimate and underestimate ETS levels, hence nicotine may not be the most appropriate



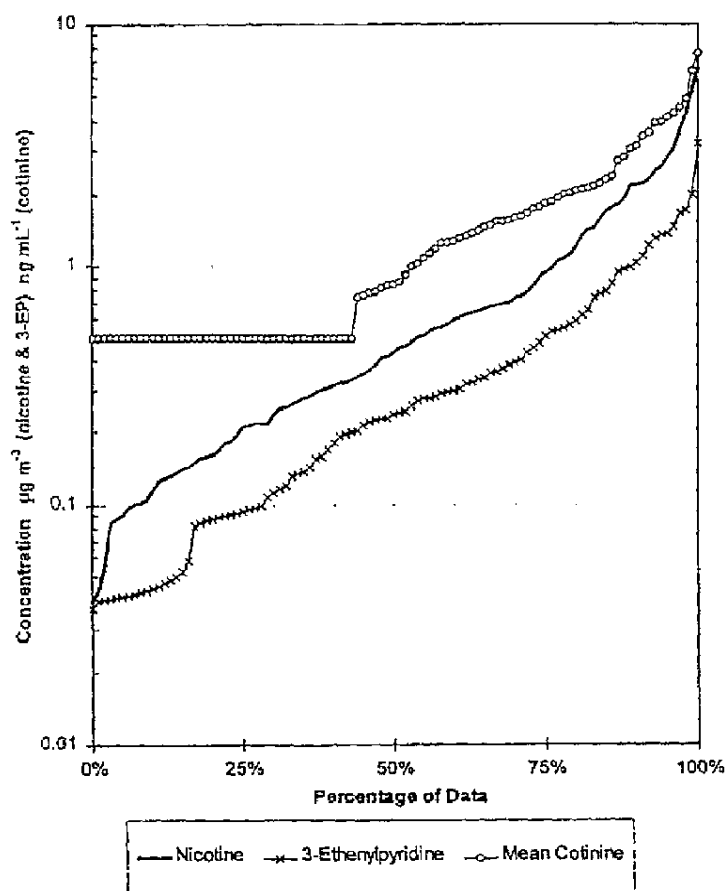


Fig. 4. Cumulative frequency distributions of average 24-h vapour phase concentrations and mean saliva cotinine concentrations (Paris).

airborne marker. It is generally believed that 3-EP is a better marker for ETS particles than nicotine, having similar decay characteristics to other ETS components and not having the strong sorptive properties of nicotine. In this study, 18% of the samples where the SolPM estimates were below the LOQ had quantifiable 3-EP levels (range 0.09–0.97  $\mu\text{g m}^{-3}$ , median 0.27  $\mu\text{g m}^{-3}$ ).

Figure 3 would suggest the expected trend of  $\text{UVPM} > \text{FPM} > \text{SolPM}$ , and shows a close similarity between the three estimates above approximately 10  $\mu\text{g m}^{-3}$ . The divergence of the lines below this level may be indicative of non-tobacco related background, resulting in an overestimation of ETS particles by UVPM and FPM. This does not, however, explain the low/undetected SolPM estimates when nicotine or 3-EP are present.

The determined nicotine and 3-EP concentrations showed good correlation with an  $R^2$  value of 0.802 and

an intercept close to zero, suggesting no bias in either measurement. Correlations for these vapour phase analytes with particulate matter estimates were moderate with  $R^2$  values in the region of 0.55. The gradient of the regression line for 3-EP vs. nicotine (Table 5) indicates nicotine levels more than twice those for the corresponding 3-EP concentrations, resulting in 38% of the determined 3-EP concentrations falling below the LOQ compared with only 5.2% for nicotine. Although both nicotine and 3-EP concentrations have been reported, subsequent exposure calculations have used nicotine values due to the lack of mainstream data for 3-EP and for ease of comparison with previous studies.

Table 5 shows the correlations for post-cotinine concentrations with both vapour and particle phase ETS components to be weak, the best correlation apparent when post-cotinine levels were compared with nicotine ( $R^2=0.196$ ). However, Fig. 4 shows that, where cotinine

Table 7. Summary statistics for 24-h TWA particle concentrations for all subjects (Paris).

Analyte	Subject group	Number of subjects	10th percentile	90th percentile	Arithmetic mean	Geometric mean	Median
RSP ( $\mu\text{g m}^{-3}$ )	Cell 1	51	29	130	71	61	62
	Cell 2	43	14	84	44	33	36
	Cell 3	45	27	120	75	65	80
	Cell 4	12	36	90	60	54	64
	Cell 5	57	20	71	44	39	43
	Cell 6	10	21	71	42	37	35
SolPM ( $\mu\text{g m}^{-3}$ )	Cell 1	48	0.39	44	13	2.9	2.7
	Cell 2	43	0.11	2.3	1.3	0.47	0.52
	Cell 3	44	1.6	45	18	8.7	9.8
	Cell 4	12	0.72	24	10	5.2	9.9
	Cell 5	57	0.33	9.3	4.2	1.9	1.9
	Cell 6	10	0.20	6.1	1.7	0.56	0.36
FPM ( $\mu\text{g m}^{-3}$ )	Cell 1	49	2.6	42	16	7.5	5.8
	Cell 2	43	0.44	4.5	2.6	1.9	2.3
	Cell 3	44	4.3	39	18	13	15
	Cell 4	12	3.1	15	10	8.0	11
	Cell 5	57	2.2	13	6.8	5.4	5.7
	Cell 6	10	1.4	10	4.7	3.5	3.0
UVPM ( $\mu\text{g m}^{-3}$ )	Cell 1	49	2.9	38	14	8.2	7.2
	Cell 2	43	0.97	6.1	3.2	2.5	2.6
	Cell 3	45	4.4	39	18	14	15
	Cell 4	12	3.3	16	10	8.8	12
	Cell 5	57	2.1	13	6.8	5.5	6.5
	Cell 6	10	1.8	10	4.7	3.6	2.7

Cell 1: smoking household; Cell 2: nonsmoking household; Cell 3: smoking household/smoking workplace; Cell 4: smoking household/nonsmoking workplace; Cell 5: nonsmoking household/smoking workplace; Cell 6: nonsmoking household/nonsmoking workplace.

Time-weighted average (TWA) exposure concentrations, determined for each subject from measured levels both inside and outside the workplace, were used to calculate the above statistical parameters for Cells 3 to 6.

levels were measurable, the distribution of concentrations was comparable to those for both nicotine and 3-EP.

#### *Concentrations of ETS constituents to which Paris subjects were exposed*

Tables 7 and 8 show summary analytical data for all subjects by Cell with the corresponding cumulative frequency distributions for SolPM, FPM, and nicotine depicted in Figs. 5, 6, and 7. The significance of any concentration differences between Cells was examined using the Wilcoxon rank sum test. Prior to the application of this nonparametric test, the Kruskal-Wallis

nonparametric analysis of variance (ANOVA) was applied to the data in order to detect if there was an overall difference between the Cells. If the overall Kruskal-Wallis analysis proved nonsignificant ( $p > 0.05$ ), any significances detected using the Wilcoxon rank sum test were considered to be false positives. For all the analytes investigated, the Kruskal-Wallis ANOVA provided evidence of a significant overall difference between Cells and subsequent pairwise comparisons of Cells were performed using the Wilcoxon rank sum test (Table 9).

The highest median RSP concentration found in this study ( $80 \mu\text{g m}^{-3}$ ) was for workers living in smoking households and working in smoking workplaces (Cell 3)

Table 8. Summary statistics for cotinine and 24-h TWA nicotine and 3-EP concentrations for all subjects (Paris).

Analyte	Subject group	Number of subjects	10th percentile	90th percentile	Arithmetic mean	Geometric mean	Median
Nicotine ( $\mu\text{g m}^{-3}$ )	Cell 1	48	0.16	2.4	0.93	0.55	0.52
	Cell 2	42	0.05	0.29	0.18	0.13	0.13
	Cell 3	41	0.50	4.1	1.8	1.3	1.4
	Cell 4	11	0.17	1.2	0.78	0.55	0.72
	Cell 5	58	0.19	1.1	0.58	0.45	0.42
	Cell 6	7	0.20	0.42	0.30	0.29	0.27
3-EP ( $\mu\text{g m}^{-3}$ )	Cell 1	48	0.10	1.3	0.49	0.30	0.30
	Cell 2	42	0.04	0.13	0.08	0.06	0.05
	Cell 3	41	0.22	1.7	0.84	0.62	0.66
	Cell 4	11	0.09	0.80	0.49	0.34	0.46
	Cell 5	58	0.09	0.53	0.29	0.23	0.24
	Cell 6	7	0.09	0.24	0.14	0.13	0.10
Cotinine* ( $\text{ng mL}^{-1}$ )	Cell 1	51	0.50	4.2	2.0	1.3	1.3
	Cell 2	44	0.50	1.4	0.75	0.62	0.50
	Cell 3	45	0.50	3.3	1.8	1.5	1.6
	Cell 4	13	0.58	3.1	1.8	1.5	1.6
	Cell 5	59	0.50	2.1	1.2	0.90	0.78
	Cell 6	10	0.50	1.3	0.70	0.62	0.50

Cell 1: smoking household; Cell 2: nonsmoking household; Cell 3: smoking household/smoking workplace; Cell 4: smoking household/nonsmoking workplace; Cell 5: nonsmoking household/smoking workplace; Cell 6: nonsmoking household/nonsmoking workplace.

3-EP=3-ethenylpyridine.

\* Values calculated from the average of pre- and post-monitoring saliva cotinine concentrations.

TWA exposure concentrations, determined for each subject from measured levels both inside and outside the workplace, were used to calculate the above vapour phase statistical parameters for Cells 3 to 6.

with an ETS particle contribution of  $9.8 \mu\text{g m}^{-3}$  (12.3%) based upon SolPM measurements. These levels, however, were not significantly different ( $p > 0.05$ ) than for workers living in smoking households and working in nonsmoking workplaces (Cell 4) with median levels of  $64 \mu\text{g m}^{-3}$  and  $9.9 \mu\text{g m}^{-3}$  apparent for RSP and SolPM, respectively. Comparing these exposure concentrations with those for housewives living in smoking households (Cell 1), there was no significant difference ( $p > 0.05$ ) between measured RSP levels.

Following the statistical evaluation of each data set for ETS particles (SolPM), there was no significant ( $p > 0.05$ ) difference between Cells 1 and 4, although median levels were 3 times higher for Cell 4. Comparison of the geometric means, which may be more appropriate, indicates the highest TWA particle concentrations were evident for workers living and working with smokers (Cell 3). The highest median nicotine

concentration ( $1.4 \mu\text{g m}^{-3}$ ) was also found for Cell 3, measured levels for this Cell being significantly higher than those determined for Cells 1 ( $p \leq 0.01$ ) and 4 ( $p \leq 0.05$ ). There was no significant difference ( $p > 0.05$ ) between saliva cotinine levels determined for subjects in Cells 1, 3, and 4.

Subjects living with nonsmokers (Cells 2, 5, and 6) were subjected to significantly lower levels of ETS particles ( $p \leq 0.05$  based on SolPM) than subjects living with smokers (Cells 1, 3, and 4) with the exception of Cell 1 vs. Cell 5. This pattern was not duplicated for nicotine, with no significant differences ( $p > 0.05$ ) evident between Cells 1, 4, 5, and 6. Nicotine levels found for workers living and working with smokers (Cell 3) were significantly higher than all other Cells investigated. Conversely, the levels of nicotine determined for housewives living with nonsmokers were significantly lower than for all other Cells investigated.

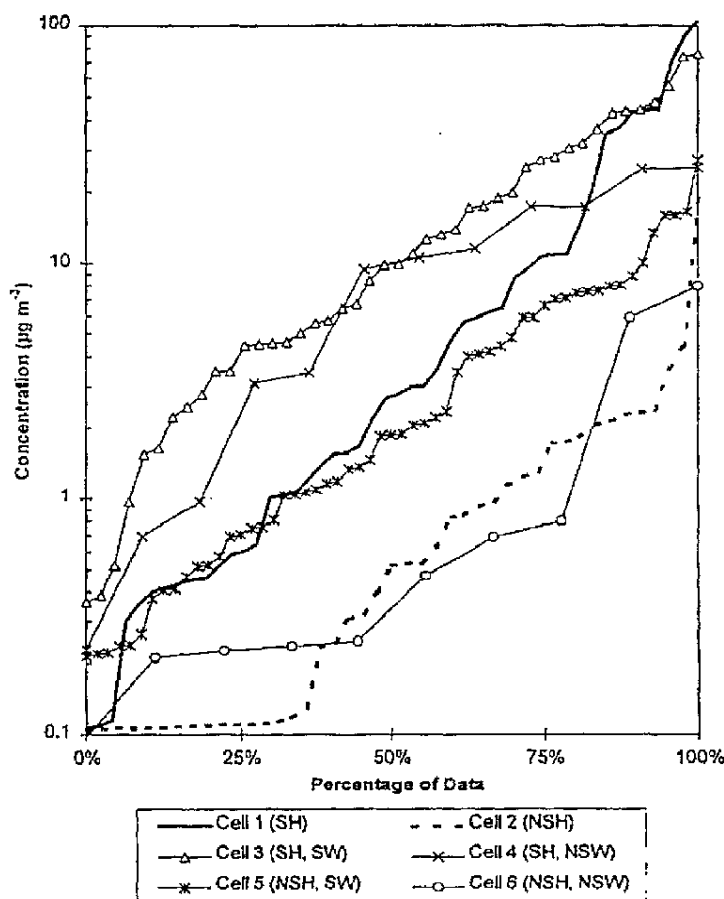


Fig. 5. Cumulative frequency distributions for SolPM by Cell (Paris).

Housewives living in nonsmoking homes encountered amongst the lowest levels of ETS found in this study, with median levels of  $36 \mu\text{g m}^{-3}$  RSP,  $0.52 \mu\text{g m}^{-3}$  ETS particles, and  $0.13 \mu\text{g m}^{-3}$  nicotine. Similar levels were found for workers both living and working in nonsmoking environments, who encountered median levels of  $35 \mu\text{g m}^{-3}$  RSP,  $0.36 \mu\text{g m}^{-3}$  ETS particles, and  $0.27 \mu\text{g m}^{-3}$  nicotine. There were no significant differences between RSP or ETS particle levels for these subjects but nicotine levels were significantly lower ( $p \leq 0.01$ ) for housewives living in nonsmoking households.

The results indicate that, for working subjects in Paris, the smoking status of the home had a major influence on their overall exposure to RSP, ETS particles, and nicotine. There was no significant difference between the levels of RSP and ETS particles found for workers residing in smoking households

irrespective of the smoking status of their workplace. However, the levels of nicotine were significantly higher for those workers who were employed in a smoking workplace. Nicotine levels found for workers residing in smoking households were also higher than those found for housewives living in smoking households.

Levels of ETS particles and nicotine, for subjects living in nonsmoking households and working in smoking workplaces (Cell 5), were not significantly different ( $p > 0.05$ ) from the levels determined for housewives living in smoking households (Cell 1). Nicotine levels were also significantly higher for subjects both living and working in smoking environments (Cell 3) than for subjects living in smoking households and working in nonsmoking workplaces (Cell 4), indicating a considerable ETS contribution from the workplace.

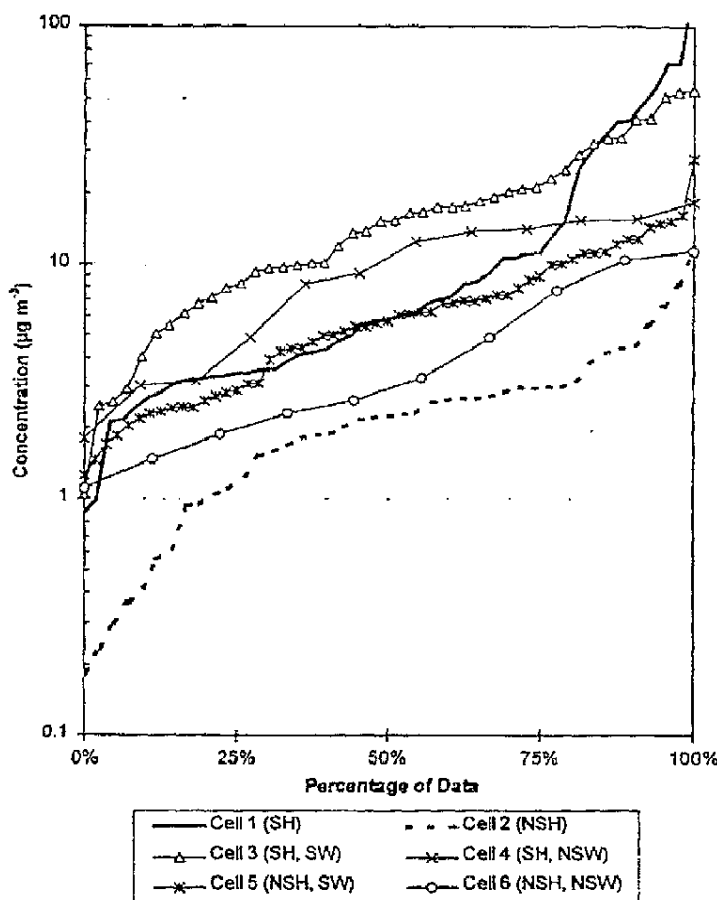


Fig. 6. Cumulative frequency distributions for FPM by Cell (Paris).

#### Saliva cotinine

Saliva cotinine concentrations, expressed as an average of pre- and post-monitoring levels, are reported by Cell in Table 8. The levels determined for subjects residing in smoking households were significantly higher than those reported for subjects living in nonsmoking households, with the highest median concentrations recorded for working subjects living in smoking households irrespective of their workplace smoking status ( $1.6 \text{ ng mL}^{-1}$ ). The lowest median levels reported were for housewives living in nonsmoking households ( $<\text{LOQ}$ ) and for working subjects both living and working in nonsmoking environments ( $<\text{LOQ}$ ). The levels reported are consistent with measured levels of ETS particles and nicotine which indicates a potential use for saliva cotinine measurements in the assessment of ETS exposure. However, a

method with a greatly improved LOQ would be required to provide, for example, the statistical significance between groups of subjects with low levels of exposure (e.g., Cell 5 and Cell 6). Cotinine measurements used for ETS exposure estimates inherit the inadequacies of nicotine behaviour and a host of other uncertainties including dietary influences, metabolic differences, and the absence of a detailed analytical method in the public domain adequate to quantify levels below  $0.1 \text{ ng mL}^{-1}$  in body fluids.

#### Exposures to RSP, ETS particles, and nicotine

Daily exposures, in terms of potential inhaled amounts ( $\mu\text{g}$ ), calculated for each Cell over the 24-h monitoring period, are summarised in Table 10. A breathing rate of  $0.65 \text{ m}^3 \text{ h}^{-1}$ , the average level of respiration calculated for awake females, was used for

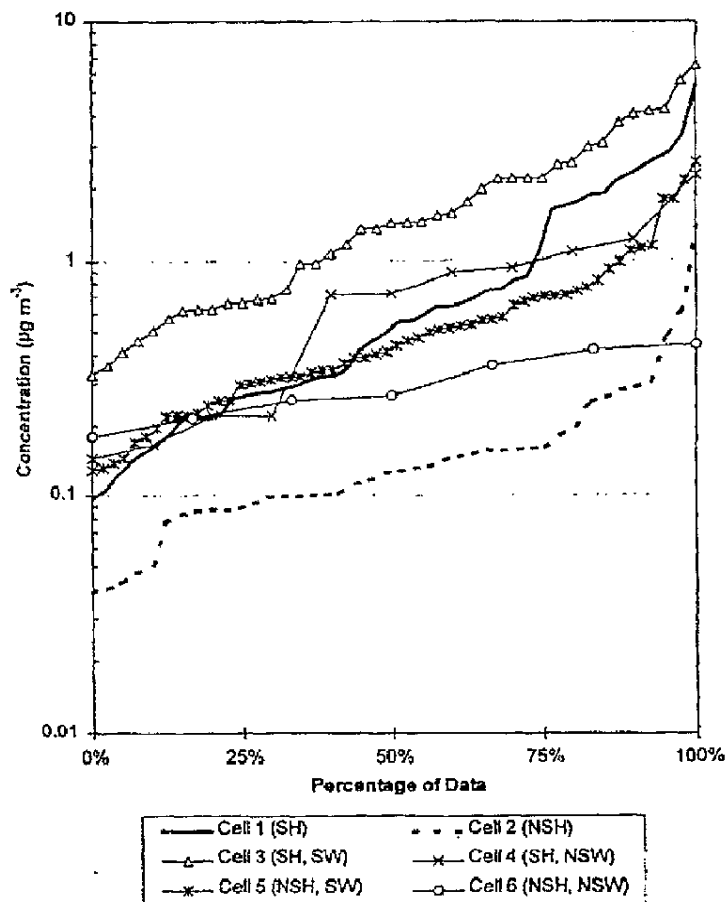


Fig. 7. Cumulative frequency distributions for nicotine by Cell (Paris).

calculating housewife exposures in Cells 1 and 2. For Cells 3 to 6, where exposures included both sexes, a breathing rate of  $0.85 \text{ m}^3 \text{ h}^{-1}$  was used, this being an average of the breathing rates for awake males ( $1.05 \text{ m}^3 \text{ h}^{-1}$ ) and females ( $0.65 \text{ m}^3 \text{ h}^{-1}$ ) as reported by Holcomb (1993). A comparable average breathing rate of  $0.93 \text{ m}^3 \text{ h}^{-1}$  was recently used by Jenkins et al. (1996) to estimate exposures on a large American study using similar personal monitoring methods to those in this study. Jenkins et al. (1996) calculated a daily nicotine intake of  $14 \text{ µg}$  for subjects either living and/or working in a smoking environment (a combination of their Cells 1, 2, and 3, equivalent to this study's Cells 3, 4, and 5). The daily exposures calculated for each Cell from the U.S. study's data are also included in Table 10. Comparing these uncombined Cells, median exposures to nicotine in Paris were similar to those determined in the U.S. for subjects living in smoking

environments irrespective of the smoking status of their workplace. However, median nicotine exposures for subjects living in nonsmoking households in the U.S. were approximately 3 to 8 times lower than those reported for Paris. The median daily exposures to RSP in Paris were approximately double those found for equivalent Cells in the U.S. study, with ETS particle exposures between 2 and 18 times higher for SolPM estimates and between 1.6 and 4 times higher for FPM estimates in Paris. In this study, the median daily exposures to ETS particles (SolPM) for working subjects living with smokers were more than 5 times higher than for those living with nonsmokers, compared with a factor of 10 for the equivalent subjects investigated by Jenkins et al. (1996) in the U.S.

The daily exposures for housewives (Cells 1 and 2) may be multiplied by 365 to provide an estimate of annual exposure, based upon both median and upper

Table 9. Significance of differences in ETS marker concentrations between Cells based upon Kruskal-Wallis ANOVA and subsequent Wilcoxon rank sum test (Paris).

SolPM		Cell 3	Cell 1	Cell 4	Cell 5	Cell 2	Cell 6
	Median	9.8	2.7	9.9	1.9	0.52	0.36
vs Cell 3	9.8	--					
vs Cell 1	2.7	**	--				
vs Cell 4	9.9	NS	NS	--			
vs Cell 5	1.9	***	NS	*	--		
vs Cell 2	0.52	***	***	***	***	--	
vs Cell 6	0.36	***	**	**	*	NS	--
FPM		Cell 3	Cell 1	Cell 4	Cell 5	Cell 2	Cell 6
	Median	15	5.8	11	5.7	2.3	3.0
vs Cell 3	15	--					
vs Cell 1	5.8	**	--				
vs Cell 4	11	NS	NS	--			
vs Cell 5	5.7	***	NS	NS	--		
vs Cell 2	2.3	***	***	***	***	--	
vs Cell 6	3.0	***	*	*	NS	NS	--
Nicotine		Cell 3	Cell 1	Cell 4	Cell 5	Cell 2	Cell 6
	Median	1.4	0.52	0.72	0.42	0.13	0.27
vs Cell 3	1.4	--					
vs Cell 1	0.52	***	--				
vs Cell 4	0.72	*	NS	--			
vs Cell 5	0.42	***	NS	NS	--		
vs Cell 2	0.13	***	***	***	***	--	
vs Cell 6	0.27	***	NS	NS	NS	**	--
Cotinine		Cell 3	Cell 1	Cell 4	Cell 5	Cell 2	Cell 6
	Median	1.6	1.3	1.6	0.78	0.50	0.50
vs Cell 3	1.6	--					
vs Cell 1	1.3	NS	--				
vs Cell 4	1.6	NS	NS	--			
vs Cell 5	0.78	***	*	*	--		
vs Cell 2	0.50	***	***	***	***	--	
vs Cell 6	0.50	***	*	**	NS	NS	--

NS: not significant ( $p > 0.05$ ); \*  $p \leq 0.05$ ; \*\*  $p \leq 0.01$ ; \*\*\*  $p \leq 0.001$ .

Cell 1: smoking household; Cell 2: nonsmoking household; Cell 3: smoking household/smoking workplace; Cell 4: smoking household/nonsmoking workplace; Cell 5: nonsmoking household/smoking workplace; Cell 6: nonsmoking household/nonsmoking workplace.

decile levels, and are presented in Table 11. This calculation assumes that the measured daily exposure is maintained throughout the year and that there is no alteration to exposure concentrations during week-ends/holidays when more time may be spent outside

the home or in the presence of the spouse. Housewives living in smoking households would be exposed annually to approximately 351 mg RSP, 15 mg ETS particles, and 3.0 mg nicotine, significantly higher than for those living in nonsmoking households who would be

Table 10. Calculated 24-h exposures to RSP, ETS particles, and nicotine assuming median and 90th percentile air concentrations (Paris).

Subject group	ETS particles			
	RSP ( $\mu\text{g}$ )	SolPM ( $\mu\text{g}$ )	FPM ( $\mu\text{g}$ )	Nicotine ( $\mu\text{g}$ )
Median levels <sup>a</sup>				
Cell 1	962	42	90	8.1
Cell 2	555	8.1	35	2.0
Cell 3	1632 (753)	200 (84)	314 (194)	29 (33)
Cell 4	1300 (522)	203 (21)	222 (87)	15 (11)
Cell 5	879 (459)	38 (2.1)	117 (34)	8.6 (2.4)
Cell 6	712 (340)	7.3 (2.1)	60 (15)	5.5 (0.7)
90th percentile levels				
Cell 1	2028	686	652	37
Cell 2	1309	37	70	4.5
Cell 3	2446	911	791	84
Cell 4	1838	498	315	25
Cell 5	1456	189	261	22
Cell 6	1440	124	214	8.6

Cell 1: smoking household; Cell 2: nonsmoking household; Cell 3: smoking household/smoking workplace; Cell 4: smoking household/nonsmoking workplace; Cell 5: nonsmoking household/smoking workplace; Cell 6: nonsmoking household/nonsmoking workplace.

NB: A breathing rate of  $0.65 \text{ m}^3 \text{ h}^{-1}$  was assumed for housewives (Cells 1-2) and  $0.85 \text{ m}^3 \text{ h}^{-1}$  for working subjects (Cells 3-6).

<sup>a</sup> Figures in brackets represent data calculated from comparable Cells reported by Jenkins et al. (1996) using their breathing rate of  $0.93 \text{ m}^3 \text{ h}^{-1}$ .

exposed to approximately 203 mg RSP, 3.0 mg ETS particles, and 0.7 mg nicotine per year. These annual exposures, based upon median levels, to ETS particles and nicotine in Paris are less than half those observed for housewives in Turin (Phillips et al. 1997b) and, for ETS particles, up to four times lower than the equivalent exposures reported for housewives in Barcelona (Phillips et al. 1997a). Based upon median levels and a typical French cigarette delivering 13 mg particles and 1 mg nicotine to the smoker, housewives living in nonsmoking households would be exposed to approximately 1 CE/y or less (FPM, SolPM, and nicotine estimates), compared with between 1.2 and 3 CE/y (SolPM and nicotine estimates, respectively) for housewives living in smoking households.

In order to estimate the annual exposures for working subjects in Cells 3 to 6, median and 90th percentile levels were calculated for each Cell from data provided by the individual monitors worn at work and outside the workplace. The annual exposures were then calculated from these values and the contributions from work and outside the workplace combined to provide exposure estimates for each Cell. To enable the calculation

of annual exposure, working subjects were assumed to have an average breathing rate of  $0.85 \text{ m}^3 \text{ h}^{-1}$  and to spend 35 h per week and 48 weeks per year at work. These calculations also assume that ETS concentrations throughout the year, both inside and outside the workplace, do not differ from those measured during the monitoring period.

Median annual exposures determined for all Cells, presented in Table 11, show the most highly exposed subjects in this study to be workers living in smoking households irrespective of the smoking status of their workplace. However, a comparison of upper decile exposures shows that the subjects in Cell 3 (smoking home, smoking workplace) were more highly exposed than the subjects in Cell 4 (smoking home, nonsmoking workplace). Based on the median levels for SolPM, FPM, and nicotine, the subjects in Cell 3 would be exposed to between 3 and 6 CE/y and between 19 and 29 CE/y, based on upper decile exposures. Subjects in Cell 4 would be exposed to approximately 6 CE/y, based on median levels, and between 10 and 14 CE/y, based on upper decile exposures. In comparison, housewives living in smoking



Table 11. Estimated annual exposures for all subjects to RSP, ETS particles, and nicotine (Paris).

Subject group	Annual exposure (mg)						
	ETS particles				Cigarette equivalents		
	RSP	SolPM	FPM	Nicotine	SolPM	FPM	Nicotine
Median levels							
Cell 1	351	15	33	3.0	1.2	2.5	3.0
Cell 2	203	3.0	13	0.74	0.23	1.0	0.74
Cell 3	487	45	76	6.0	3.5	5.8	6.0
Cell 4	359	78	83	6.2	6.0	6.4	6.2
Cell 5	266	6.0	26	2.4	0.46	2.0	2.4
Cell 6	278	1.7	16	1.8	0.13	1.2	1.8
90th percentile levels							
Cell 1	740	230	238	13	19	18	13
Cell 2	478	13	26	1.7	1.0	2.0	1.7
Cell 3	938	372	241	29	29	19	29
Cell 4	736	185	147	10	14	11	10
Cell 5	546	56	75	5.4	4.3	5.8	5.4
Cell 6	509	24	52	2.9	1.8	4.0	2.9

Cell 1: smoking household; Cell 2: nonsmoking household; Cell 3: smoking household/smoking workplace; Cell 4: smoking household/nonsmoking workplace; Cell 5: nonsmoking household/smoking workplace; Cell 6: nonsmoking household/nonsmoking workplace.

NB: A breathing rate of  $0.65 \text{ m}^3 \text{ h}^{-1}$  was assumed for housewives (Cells 1-2) and  $0.85 \text{ m}^3 \text{ h}^{-1}$  for working subjects (Cells 3-6). Annual exposures for housewives were calculated by simple extrapolation of their 24-h exposure levels. Annual exposures were calculated for working subjects assuming a 35-h working week and 48-week working year with the remainder of the time spent at "home". Median and 90th percentile concentrations of ETS markers for workers at "work" and at "home" were calculated from the data provided by individual monitors for each Cell.

households (Cell 1) would be exposed to between 1 and 3 CE/y, based on median levels, and between 13 and 19 CE/y for the most highly exposed, based on upper decile levels.

Applying the same criteria used for the calculations in Table 11, it was possible to estimate the contribution of the workplace to overall annual exposure. These have been expressed as a percentage of both median and upper decile results and are summarised in Table 12. At the median level, workplace ETS particle contributions vary from approximately 7% in Cell 4 to 79% in Cell 5, based upon SolPM measurements, and between 12% and 50% for the same Cells, respectively, based upon FPM. In this instance, ETS particle contributions based upon FPM determinations more accurately reflected the workplace contributions of nicotine to annual exposure. Overall, the workplace contributes between 33% and 51% of annual exposures to ETS particles and nicotine.

#### *Concentrations of RSP, ETS particles, nicotine, and 3-EP by location*

The magnitude of exposures to RSP, ETS particles, and nicotine for working subjects, both inside and outside of the workplace, were assessed using data provided by the individual monitors. The individual monitor contributions were combined to provide an estimate of exposure concentrations in smoking (Table 13) and nonsmoking (Table 14) environments, both inside and outside the workplace. The comparison of saliva cotinine levels was not meaningful in this case and the data was excluded from the tables.

As would be expected, median levels of RSP, ETS particles, and nicotine were found to be higher in smoking environments than in nonsmoking environments. Based on the median levels reported in Table 13, the levels of ETS encountered in the smoking workplace were higher than those encountered outside the

Table 12. Estimated contribution of the workplace to annual exposures of RSP, ETS particles, and nicotine for all working subjects (Paris).

Subject	ETS particles			
group	RSP	SolPM	FPM	Nicotine
Median levels				
Cell 3	23%	20%	25%	33%
Cell 4	21%	7%	12%	7%
Cell 5	31%	79%	50%	51%
Cell 6	26%	35%	30%	33%
Overall <sup>a</sup>	26%	51%	39%	45%
90 <sup>th</sup> percentile levels				
Cell 3	25%	31%	34%	29%
Cell 4	18%	11%	13%	18%
Cell 5	27%	84%	63%	63%
Cell 6	22%	92%	40%	30%
Overall <sup>a</sup>	24%	33%	33%	42%

Cell 3: smoking household/smoking workplace; Cell 4: smoking household/nonsmoking workplace; Cell 5: nonsmoking household/smoking workplace; Cell 6: nonsmoking household/nonsmoking workplace.

<sup>a</sup> Cells 3 to 6 combined.

Table 13. Summary analytical statistics for employed subjects in smoking environments (Paris).

Analyte	Environment <sup>a</sup>	Number of subjects	10th percentile	90th percentile	Arithmetic mean	Geometric mean	Median
RSP ( $\mu\text{g m}^{-3}$ )	work	104	16	127	71	56	63
	home	58	26	112	66	55	61
SolPM ( $\mu\text{g m}^{-3}$ )	work	104	0.38	49	16	4.6	3.8
	home	57	0.22	39	14	4.8	6.5
FPM ( $\mu\text{g m}^{-3}$ )	work	104	2.5	44	18	11	12
	home	57	1.7	25	13	8.1	9.5
UVPM ( $\mu\text{g m}^{-3}$ )	work	104	3.0	45	18	12	12
	home	58	2.0	28	14	8.6	9.0
Nicotine ( $\mu\text{g m}^{-3}$ )	work	102	0.33	4.6	1.9	1.1	1.0
	home	56	0.24	2.9	1.2	0.72	0.68
3-EP ( $\mu\text{g m}^{-3}$ )	work	102	0.13	2.1	0.87	0.55	0.59
	home	56	0.08	1.4	0.61	0.37	0.35

<sup>a</sup> work - data from the "workplace" monitor of subjects in Cells 3 and 5;

home - data from the "home" monitor of subjects in Cells 3 and 4.

workplace for those subjects living in smoking homes. The exception to this general rule was the median level for SolPM which was higher outside the workplace ( $6.5 \mu\text{g m}^{-3}$ ) than inside ( $3.8 \mu\text{g m}^{-3}$ ). This may be due to the larger numbers of determinations which yielded

results below the LOQ for SolPM. The median levels determined for ETS particle and vapour phase components in nonsmoking environments were more consistent, again levels being higher in the workplace than those outside the workplace. Comparing the home data

Table 14. Summary analytical statistics for employed subjects in nonsmoking environments (Paris).

Analyte	Environment <sup>a</sup>	Number of subjects	10th percentile	90th percentile	Arithmetic mean	Geometric mean	Median
RSP ( $\mu\text{g m}^{-3}$ )	work	22	18	93	53	45	53
	home	67	10	66	36	30	31
SolPM ( $\mu\text{g m}^{-3}$ )	work	22	0.27	15	5.0	1.5	1.2
	home	67	0.16	1.4	0.81	0.34	0.20
FPM ( $\mu\text{g m}^{-3}$ )	work	22	1.7	14	7.4	5.1	4.1
	home	67	0.84	4.6	3.0	2.2	2.2
UVP ( $\mu\text{g m}^{-3}$ )	work	22	2.4	14	8.1	5.9	5.7
	home	67	0.94	4.9	2.9	2.3	2.1
Nicotine ( $\mu\text{g m}^{-3}$ )	work	20	0.13	0.99	0.51	0.36	0.33
	home	65	0.13	0.34	0.23	0.19	0.19
3-EP ( $\mu\text{g m}^{-3}$ )	work	20	0.10	0.68	0.27	0.19	0.14
	home	65	0.06	0.18	0.10	0.09	0.07

<sup>a</sup> work - data from the "workplace" monitor of subjects in Cells 4 and 6;

home - data from the "home" monitor of subjects in Cells 5 and 6.

Table 15. Estimated annual exposures to RSP, ETS particles, and nicotine by environment (Paris).

		Annual exposure (mg)						
		ETS particles				Cigarette equivalents		
Environment		RSP	SolPM	FPM	Nicotine	SolPM	FPM	Nicotine
Median levels								
SM	home	367	39	57	4.1	3.0	4.4	4.1
NS	home	184	1.2	13	1.1	0.09	1.0	1.1
SM	work	90	5.4	16	1.4	0.42	1.2	1.4
NS	work	76	1.7	5.9	0.47	0.13	0.45	0.47
90th percentile levels								
SM	home	671	233	152	18	18	12	18
NS	home	398	8.3	28	2.0	0.64	2.2	2.0
SM	work	182	70	63	6.6	5.4	4.8	6.6
NS	work	132	22	20	1.4	1.7	1.5	1.4

SM - smoking environment; NS - nonsmoking environment.

NB - A 35-h working week and 48-week working year was assumed for the calculation of annual exposure at work. The remainder of the time outside of the workplace was assumed to be at 'home'. An average breathing rate of  $0.85 \text{ m}^3 \text{ h}^{-1}$  was assumed at all times and the median/90th percentile concentrations in the various locations were taken from the 'combined Cell' data reported in Tables 13 and 14. Overall annual exposures may be estimated by summing the data from the requisite 'home' and 'work' environments.

Table 16. Subjective assessment of the single environment where subjects considered themselves to be most exposed to ETS (Paris).

Environment	% of Responses*
Bar/restaurants	44.8
Work	20.7
Home	15.5
Outdoors	6.9
Other locations indoors	6.3
Travelling	3.4
Nowhere/not exposed	2.3

\* Calculated as a percentage of valid responses to this question, 48 subjects having failed to answer this question correctly.

in Table 13 (smoking environments) with the data presented in Tables 7 and 8 for Cell 1 (housewives from smoking households), there is an indication that worker exposures outside the workplace in Paris are higher than those for housewives.

Estimations of annual exposures for workers, based on location and performed using an assumed 35-h work week and a 48-week work year, are reported in Table 15. By combining the different permutations of these annual exposures for the workplace and outside the workplace, the overall annual exposures may be estimated on a Cell by Cell basis. A comparison of these exposure combinations with the exposures actually calculated for individual Cells show close agreement. Based on the measured median levels, subjects both living in smoking households and working in smoking workplaces would be exposed to between 3.4 and 5.6 CE/y (3.5 and 6.0 CE/y for Cell 3) compared to between 0.2 and 1.6 CE/y for subjects living and working in nonsmoking environments (0.1 and 1.8 CE/y for Cell 6).

#### Subjective comparisons of ETS exposure

The ETS exposures of individuals in smoking and nonsmoking environments were extensively investigated as part of this study. Information from the subjects' diaries, completed during the monitoring periods and the last visit survey questionnaires, indicated that approximately 14% of the subjects working in smoking environments did not see or smell any smoking during the monitoring period. Also significant was the fact that about 57% of all subjects working in nonsmoking environments did note smoking during the monitoring period. From analysis of the diaries and combining the single and dual monitor studies, 17% of the subjects

living in a smoking household did not note smoking taking place during the monitoring period. Conversely, 32% of the subjects living in a nonsmoking household did note smoking taking place.

As part of the last visit survey, the study subjects were asked a number of subjective questions regarding their exposure to ETS, both in general and during the 24-h monitoring period. Table 16 lists the various environments and the percentage of subjects indicating the environment where they believe most exposure to tobacco smoke takes place. Approximately 4 out of 5 subjects believed that they were most exposed to ETS outside the workplace. Also evident is the general perception that their highest exposure takes place in restaurants/bars.

#### CONCLUSIONS

In this study, the highest median concentrations of RSP were recorded for subjects living and working in smoking environments. The lowest RSP levels were found for housewives living with nonsmokers.

Exposure to ETS particles was highest for employed subjects living with smokers irrespective of the smoking status of the workplace, possibly indicating that the home has most influence on ETS exposure. Currently, solanesol appears to be the most appropriate marker for ETS particle exposure, although an improved method for its determination should be a consideration for the future.

The rate at which subjects misreported their non-smoking status varied between 1.8 and 4.7% based on saliva cotinine levels. The use of saliva cotinine as a biomarker for ETS exposure should be further investigated, to include the development of a method with a much improved LOQ.

*Acknowledgment*—The funding for this study was made available to Covance Laboratories Ltd. by the Center for Indoor Air Research (CIAR), Linthicum, MD, USA. Thanks go to research nurse Eva Götharsson at the Karolinska Institute, Göran Tamm and Christer Hermansson at MarknadsAnalys, and to the Department of Biopharmaceutical Analysis at Covance Laboratories Ltd. where all the samples were analysed.

#### REFERENCES

- Back, S.-O.; Kim, Y.-S.; Perry, R. Indoor air quality in homes, offices and restaurants in Korean urban areas - Indoor/outdoor relationships. *Atmos. Environ.* 31: 529-544; 1997.
- Benowitz, N.L.; Jacob, P.; Sachs, D.P.L. Deficient C-oxidation of nicotine. *Clin. Pharmacol. Ther.* 57: 590-594; 1995.
- Cholerton, S.; et al. Poor metabolizers of nicotine and CYP2D6 polymorphism. *Lancet* 343: 62-63; 1994.

- Davis, R.A.; Stiles, M.F. Determination of nicotine and cotinine: Comparison of GC and radioimmunoassay methods. Paper presented at the 47th Tobacco Chemists' Research Conference, Gatlinburg, Tennessee, 18-21 October 1993. Available from: R.J. Reynolds R & D Technical Services Library, PO Box 2959, Winston Salem, NC 27102.
- Delfino, R.J.; Ernst, P.; Jaakkola, M.S.; Solomon, S.; Becklake, M.R. Questionnaire assessments of recent exposure to environmental tobacco smoke in relation to salivary cotinine. *Eur. Respir. J.* 6: 1104-1108; 1993.
- Eatough, D.J. Assessing exposure to environmental tobacco smoke. In: Nagda, N.L., ed. *Modeling of indoor air quality and exposure*. ASTM STP 1205. Philadelphia, PA: American Society for Testing and Materials; 1993: 42-63.
- Etzel, R.A. A review of the use of saliva cotinine as a marker of tobacco smoke exposure. *Prev. Med.* 19: 190-197; 1990.
- Heavner, D.L.; Morgan, W.T.; Ogden, M.W. Determination of volatile organic compounds and respirable particulate matter in New Jersey and Pennsylvania homes and workplaces. *Environ. Int.* 22: 159-183; 1996.
- Holcomb, L.C. Indoor air quality and environmental tobacco smoke: Concentration and exposure. *Environ. Int.* 19: 9-40; 1993.
- Jenkins, R.A.; Palausky, A.; Counts, R.A.; Bayne, C.K.; Dindal, A.B.; Guerin, M.W. Exposure to environmental tobacco smoke in sixteen cities in the United States as determined by personal breathing zone air sampling. *J. Exp. Anal. Environ. Epidemiol.* 6: 473-502; 1996.
- Lee, P.N. Lung cancer and passive smoking: Association an artifact due to misclassification of smoking habits? *Toxicol. Lett.* 35: 157-162; 1987.
- McNeill, A.D.; Jarvis, M.J.; West, R.; Russell, M.A.H.; Bryant, A. Saliva cotinine as an indicator of cigarette smoking in adolescents. *Brit. J. Addiction* 82: 1355-1360; 1987.
- Nelson, P.R.; Heavner, D.L.; Oldaker, G.B. III. Problems with the use of nicotine as a predictive environmental tobacco smoke marker. In: *Measurement of toxic and related air pollutants: Environmental Protection Agency/Air and Waste Management Association International Symposium*. Pittsburgh, Pa: Air and Waste Management Association; 1990: 550-555.
- Nelson, P.R.; Conrad, F.W.; Kelly, S.P.; Maiolo, K.C.; Richardson, J.D.; Ogden, M.W. Composition of environmental tobacco smoke (ETS) from international cigarettes and determination of ETS-RSP: Particulate marker ratios. *Environ. Int.* 23: 47-52; 1997.
- Ogden, M.W.; Eudy, J.W.; Heavner, D.L.; Conrad, F.W.; Green, C.R. Improved gas chromatographic determination of nicotine in environmental tobacco smoke. *Analyst* 114: 1005-1008; 1989.
- Ogden, M.W.; Maiolo, K.C.; Oldaker, G.B.; Conrad, F.W. Evaluation of methods for estimating the contribution of ETS to respirable suspended particles. In: Walkinshaw, D.S., ed. *Indoor air 90 2*. International conference on indoor air quality and climate. Ottawa 1990. 1990: 415-420. Available from: Canada Mortgage and Housing Corp., Ottawa, Ontario.
- Ogden, M.W.; Heavner, D.L.; Foster, T.L.; Maiolo, K.C.; Cash, S.L.; Richardson, J.L.; Martin, P.; Simmons, P.S.; Conrad, F.W.; Nelson, P.R. Personal monitoring system for measuring environmental tobacco smoke exposure. *Environ. Technol.* 17: 239-250; 1996.
- Ogden, M.W.; Morgan, W.T.; Heavner, D.L.; Davis, R.A.; Steichen, T.J. National incidence of smoking and misclassification among the U.S. married female population. *J. Clin. Epidemiol.* 50: 253-263; 1997.
- Phillips, K.; Howard, D.A.; Browne, D.; Lewsley, J.M. Assessment of personal exposures to environmental tobacco smoke in British nonsmokers. *Environ. Int.* 20: 693-712; 1994.
- Phillips, K.; Bentley, M.C.; Howard, D.A.; Alván, G. Assessment of air quality in Stockholm by personal monitoring of nonsmokers for respirable suspended particles and environmental tobacco smoke. *Scand. J. Work Environ. Health* 22 Suppl. 1: 1-24; 1996.
- Phillips, K.; Bentley, M.C.; Howard, D.A.; Alván, G.; Huici, A. Assessment of air quality in Barcelona by personal monitoring of nonsmokers for respirable suspended particles and environmental tobacco smoke. *Environ. Int.* 23: 173-196; 1997a.
- Phillips, K.; Howard, D.A.; Bentley, M.C.; Alván, G. Assessment of air quality in Turin by personal monitoring of nonsmokers for respirable suspended particles and environmental tobacco smoke. *Environ. Int.* 23: 851-871; 1997b.
- Sterling, E.M.; Collett, C.W.; Ross, J.A. Assessment of nonsmokers' exposure to environmental tobacco smoke using personal-exposure and fixed-location monitoring. *Indoor Built Environ.* 5: 112-125; 1996.
- Wagenknecht, L.E.; Burke, G.L.; Perkins, L.L.; Haley, N.J.; Friedman, G.D. Misclassification of smoking status in the CARDIA study: A comparison of self-report with serum cotinine levels. *Am. J. Public Health* 82: 33-36; 1992.
- Van Vunakis, H.; Gjika, H.B.; Lagone, J.J. Method 16 - Radioimmunoassay for nicotine and cotinine. In: O'Neill, I.K.; Brunnemann, K.D.; Dodet, B.; Hoffman, D., eds. *Environmental carcinogens methods of analysis and exposure measurement*. 9: 317-330; 1987.